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Contract Description

The purpose of this work is to develop techniques to overcome the fundamental limits of present methods for high resolution spectroscopy and frequency standards--the second order and residual first-order Doppler shifts. To this end, we study suitable frequency reference transitions in ions which are stored in electromagnetic traps and cooled by radiation pressure to $< 1\text{K}$.

Scientific Problem

The scientific problems are (1) to suppress second order and residual first order Doppler shifts in atomic frequency standards in a fundamental way--by substantially reducing the kinetic energy of ions stored ion electro-magnetic traps, (2) to study suitable reference transitions in ions that can be used as frequency standards, and (3) to study the problems generic to all stored ion frequency standards. The goal is to achieve at least a factor of 100 improvement in accuracy over the present best device, the Cesium beam frequency standard, which has an accuracy of approximately 2 parts in 10^{14} .

Scientific and Technical Approach

Laser cooling is employed on all experiments in order to suppress Doppler shifts. Temperatures $< 0.1\text{K}$ are routinely achieved. To avoid light shifts on "clock" transitions we investigate "sympathetic cooling" where one ion species is laser cooled and by Coulomb collisions cools another ion species of spectroscopic interest. We will continue experiments on Mg^+ and Be^+ in order to study generic problems with traps. We are developing a separate experiment for Hg^+ ions. These experiments have the goal of realizing a frequency standard with 10^{-15} or better accuracy.

Summary of progress since Oct., 1986

(1) Optical Sideband Cooling and the Fundamental Limit of Laser Cooling Achieved.

The fundamental limit of cooling for any particle which is contained by some apparatus (the electrodynamic trap in our experiment, or the walls of the vacuum apparatus in a neutral atom experiment) is putting the atom in the zero point state of motion. We have achieved this (for the first time) using single Hg^+ ions.

(2) Precise Test of Quantum Jump Theory Completed.

An experiment, designed around a single Mg^+ ion in a Penning trap was completed and compared with theory. This is the only high precision comparison of theory and experiment and agreement is found to within the experimental uncertainty. (2%).

(3) Photon Antibunching and Subpoissonian Statistics.

These characteristics of the $11\ \mu\text{m}\ 2\text{P}_{3/2} \rightarrow 2\text{D}_{3/2}$ photon emission in a single Hg^+ ion have been detected via the quantum jumps of the 194 nm fluorescence from this ion.

(4) Lifetimes and Branching Ratios in Hg^+ .

Lifetimes of the $2\text{P}_{1/2}$, $2\text{D}_{3/2}$, $2\text{D}_{5/2}$ levels and branching ratios from the $2\text{P}_{1/2}$ and $2\text{D}_{3/2}$ levels are obtained solely from the statistics of quantum jumps on the $2\text{S}_{1/2} \rightarrow 2\text{P}_{1/2}$ transition.

- (5) Laser Cooling in Penning Traps. The theory of laser cooling in Penning traps (complicated by the unstable rotation degree of freedom) was completed and found to agree with our experimental results.
- (6) Laser Development. Nd:FAP made to lase single frequency. This laser has a frequency one-fourth that of the optical "clock" transition in Hg^+ . Since it is a solid state laser it is intrinsically more stable than the dye lasers currently used and will be quadrupled for optical clock use.
- (7) Coulomb Clusters, Liquid and Solid Plasmas. Since the limiting systematic uncertainty in the spectroscopic experiments is still expected to be due to Doppler frequency shifts, it is critical to understand the ion distribution functions. As an outgrowth of these studies we have studied the spatial correlations of laser cooled ions in both Paul (rf) and Penning traps.
- (8) Synchrotron Frequency Divider. Superconducting magnet tested. New trap design nearly complete.
- (9) Sympathetic Cooling. Achieved in superconducting magnet trap with Mg^+ and Be^+ ions. This is the first important step in achieving much higher resolution on hyperfine structure than obtained previously.
- (1) Optical Sideband Cooling and the Fundamental Limit of Laser Cooling Achieved. In all laser-cooling experiments performed to date, cooling has been accomplished in the regime where the oscillation or vibration frequency ω_v of the particle in its confining well was less than the natural width γ of the cooling transition. This regime also applies to "free" atom cooling experiments such as optical molasses ($\omega_v \rightarrow 0$). In recent preliminary experiments by us, laser-cooling in the sideband limit (where $\omega_v > \gamma$) was accomplished for the first time. Using this technique, a single trapped Hg^+ ion was cooled such that it spent most of its time in the ground state quantum level of the confining potential. To the extent that the ion is in the zero point energy state of motion, this realizes for the first time the fundamental limit of laser cooling and the ideal of an isolated atomic particle at rest in free space to



within the quantum mechanical limits imposed by the surrounding apparatus.

The ion was confined in a Paul(rf) trap and first cooled to near the "Doppler cooling limit" of $T = \hbar\gamma/2k_B \approx 1.7\text{mK}$ by scattering 194 nm photons on the strong $^2S_{1/2} - ^2P_{1/2}$ dipole transition. In a second step, the 194 nm radiation was switched off and the $^2S_{1/2} - ^2D_{5/2}$ quadrupole transition at 282 nm was driven on the resolved first lower secular motion sideband frequency by a narrowband dye laser with an intensity of approximately 10 W/cm^2 . This radiation was derived from a frequency doubled dye laser. The radiation bandwidth at 282 nm was approximately 40 kHz. In order to enhance the speed of the sideband cooling, the 90 ms lifetime of the $^2D_{5/2}$ state was reduced by driving the $^2D_{5/2} - ^2P_{3/2}$ transition ($\lambda = 398\text{ nm}$) by another frequency doubled dye laser. From the $^2P_{3/2}$ state, the ion rapidly decays to the $^2S_{1/2}$ ground state. The kinetic energy of the ion after cooling was probed by measuring the excitation probability of the $^2S_{1/2} - ^2D_{5/2}$ transition at the carrier and at the sideband frequencies.

In a first experiment, the potential well was nonspherical and only the axial motion at $\omega_v/2\pi = 4\text{ MHz}$ was cooled below the Doppler cooling limit. After cooling, the excitation rate on the first upper sideband (Stokes transition) at frequency $\omega_0 + \omega_v$ was three times stronger than on the first lower (anti-Stokes) sideband (Fig. 1). This indicates that the ion is in the ground state ($n_v = 0$) level of motion with approximately 2/3 probability. The kinetic energy of the axial motion corresponds to a temperature of about $175\text{ }\mu\text{K}$.

In a second experiment, the ion was kept in an approximately spherical potential well (secular frequencies $\approx 2.4\text{ MHz}$) and all degrees of freedom were cooled to near 1.7 mK by two orthogonal beams of 194 nm radiation. From the fixed spatial orientation along the "x" axis of two laser cooled ions in the trap it is known that the radial frequencies are nondegenerate. Thus a single 282 nm beam directed at an angle so that it is not perpendicular to any of the trap axes and overlapped in frequency

with the first lower nondegenerate sidebands cools all degrees of freedom simultaneously.

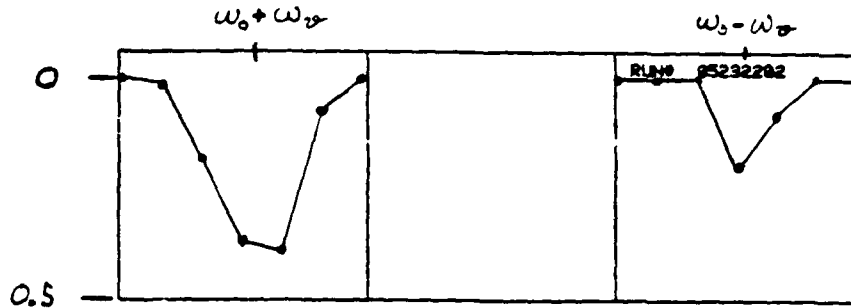


Fig. 1. Probability of absorption (vertical axis) on the lower (right) and upper (left) sidebands of the quadrupole transitions in Hg^+ after sideband cooling. The imbalance indicates the ion spends most of its time in the zero point energy state of motion.

(2) Precise test of Quantum Jump Theory Completed

We have observed quantum jumps in an atomic system which is quite different than those in which quantum jumps had been previously observed: a single laser is used to induce spontaneous Raman transitions into and out of a ground state shelving level. This new system has particular advantages for precisely testing the statistical predictions of the quantum jump theory. The process is illustrated in Fig. 2. The single radiation source is tuned between a pair of Zeeman levels of an atom or ions with a $^2S_{1/2}$ ground state and an excited $^2P_{3/2}$ state. The six Zeeman sub-levels in Fig. 2 are labelled from 1 to 6 for convenience. The frequency ω of the radiation is tuned near the level 1 \leftrightarrow level 3 transition frequency ω_0 . The atom cycles nearly continuously between these levels since the dipole selection rules allow spontaneous decay only to the original ground level. A steady stream of fluorescence photons, which are readily detected, is emitted by the atom during this period. However, if the radiation is linearly polarized perpendicular to the direction of the magnetic field, the 1 \leftrightarrow 5 transition is also allowed, although it is far from resonance. This transition is indicated by the dashed arrow in Fig. 2. A spontaneous decay from level 5 can then leave the atom in the "other" ground level (i.e. level 2). This spontaneous Raman transition into level 2 takes the atom out of the 1 \leftrightarrow 3 cycling loop, causing the emitted fluorescence to

suddenly stop. The off-resonant $2 \rightarrow 4 \rightarrow 1$ spontaneous Raman transition (not shown in Fig. 2) will return the atom to the cycling loop where it will resume scattering. Consequently, the detected fluorescence will alternate between periods of "on" and "off".

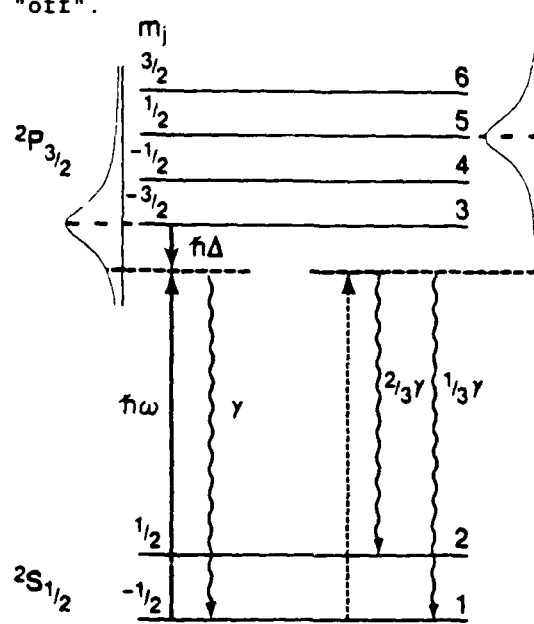


Figure 2. The energy level structure of the $2S_{1/2}$ and the $2P_{3/2}$ states of an atom in a magnetic field. Off-resonant excitations from level 2 and spontaneous decay from levels 4 and 6 are not shown.

We have used a single $^{24}\text{Mg}^+$ ion confined by the static magnetic and electric fields of a Penning trap to demonstrate this process. The magnetic field in the trap was 1.39 T, which gives $\alpha \approx 1200\gamma$, where $\hbar\alpha/2$ is the energy separation between adjacent excited state sublevels and $\gamma = (2\pi)43$ MHz is the excited state spontaneous decay rate (see Fig. 2). The $3S_{1/2}$ to $3P_{3/2}$ transition wavelength is 280 nm for which we generated up to 200 μW by frequency doubling the output of a dye laser. The fluorescence photons were collected in a direction perpendicular to both the magnetic field and laser beam directions. Because of the large collection solid angle, we were able to detect up to 2×10^5 photons/s.

We have used the density-matrix formulation to determine the dynamics of the population evolution among the six levels for this system. The system of

equations consists of the six population equations for ρ_{11} - ρ_{66} and for σ polarized radiation, there are six nonzero coherences σ_{13} , σ_{15} , σ_{24} , σ_{26} , ρ_{35} and ρ_{46} . The four σ_{ij} are due to direct laser coupling of a ground and an excited level, while the two ρ_{ij} represent the stimulated Raman coherences between pairs of excited levels. Upon solving the equations we found that when $\alpha \gg \gamma, \Omega, \Delta$ (Ω is the Rabi frequency of the $1 \leftrightarrow 3$ transition) the evolution for times longer than γ^{-1} is governed by the rates in which population is transferred between the levels participating in the strongly fluorescing cycling and those which do not.

Because of the off-resonance nature of the transitions between these systems, these rates do not show the effect of saturation of the $1 \leftrightarrow 3$ transition. Specifically, the stimulated Raman coherence ρ_{35} mediates population transfer between the excited levels 3 and 5 which compensates for the decrease of population in level 1 with increasing intensity. Also, because the ratio of average on to off times $\langle T_{on} \rangle / \langle T_{off} \rangle = 16 + O(\Delta/\alpha)$ is largely independent of the experimental parameters (laser intensity, detuning and magnetic field strength) we can perform a quantitative, precise test of the theory. Table I shows a comparison of experiment with theory for seven data runs, each with a different value of the Rabi frequency (i.e. laser intensity). The theory which does not include the ρ_{35} coherence predicts that $\langle T_{on} \rangle / \langle T_{off} \rangle$ will range between 16 for $\Omega \ll \gamma$ and 32 for $\Omega \gg \gamma$. Our data verifies the complete theory, which includes the effects of coherences between excited states, to a precision of 2%. This work is detailed in ref. 6 of "Papers published."

(3.) Photon Antibunching and Subpoissonian Statistics

Antibunching and sub-Poissonian statistics of the 11 μm photon emission from the $5d^{10}6p \ ^2P_{1/2}$ to $5d^96s^2 \ ^2D_{3/2}$ transition in one and two trapped Hg^+ ions have been detected indirectly from the interruptions (quantum jumps) of the laser-induced-fluorescence of the 194 nm $5d^{10}6s \ ^2S_{1/2}$ to $5d^{10}6p \ ^2P_{1/2}$ first resonance transition.

Hg^+ ions confined in a Paul (rf) trap were irradiated with narrowband, cw 194 nm radiation. The 194 nm photons emitted by the ions were detected by a photomultiplier tube. About once in 10^7 times the $^2P_{1/2}$ level decayed to the

metastable (9 ms lifetime) $^2D_{3/2}$ level with the emission of an 11 μm photon, instead of to the ground $^2S_{1/2}$ level. When this happened, the 194 nm

Table I

RUN	Ω^2/γ^2	$-\Delta/\gamma$	CALCULATED		MEASURED
			$\frac{\langle T_{on} \rangle}{\langle T_{off} \rangle} \left(\frac{w/o}{c_{oh.}} \right)$	$\frac{\langle T_{on} \rangle}{\langle T_{off} \rangle} \left(\frac{w}{c_{oh.}} \right)$	$\frac{\langle T_{on} \rangle}{\langle T_{off} \rangle}$
1	0.65 ± 0.09	0.67 ± 0.03	19.1 ± 0.2	16.09	15.76 ± 0.84
2	1.82 ± 0.16	$0.22 \pm \begin{smallmatrix} 0.10 \\ -0.20 \end{smallmatrix}$	25.8 ± 0.5	16.03	16.02 ± 0.67
3	1.89 ± 0.16	$0.20 \pm \begin{smallmatrix} 0.12 \\ -0.20 \end{smallmatrix}$	26.1 ± 0.6	16.03	15.52 ± 0.58
4	2.02 ± 0.16	0.84 ± 0.05	21.7 ± 0.3	16.11	16.58 ± 0.59
5	2.45 ± 0.25	1.07 ± 0.05	21.0 ± 0.2	16.13	16.13 ± 0.70
6	2.61 ± 0.20	0.60 ± 0.05	24.4 ± 0.6	16.08	17.35 ± 0.70
7	2.64 ± 0.16	0.50 ± 0.06	25.2 ± 0.6	16.07	16.26 ± 0.69

fluorescence of that ion suddenly ceased. After the ion reached the ground state, either by decaying directly from the $^2D_{3/2}$ level or by first decaying to the metastable (90 ms lifetime) $5d^96s^2 \ ^2D_{3/2}$ level, its 194 nm fluorescence suddenly returned to its previous level. When a single ion was trapped, the fluorescence was observed to switch spontaneously between a steady level and zero. Each transition from the fluorescence-on state to the fluorescence-off state was assumed to mark the emission of an 11 μm photon. Data were also obtained with two ions, separated by about 3 μm . In that case, the fluorescence switched between three values, corresponding to 0, 1, or 2 of the ions being in a metastable level, and each transition from a higher to a lower intensity of fluorescence was assumed to mark the emission of an 11 μm photon from one of the ions.

Let $g^{(2)}(\tau)$ be the normalized intensity correlation function of the 11 μm field. Photon antibunching is said to occur if $g^{(2)}(0) < 1$. Figure 3 shows

plots of $g^{(2)}(\tau)$ for (a) one ion and (b) two ions. The solid curves are calculated from density-matrix equations. Both the one-ion and two-ion plots show antibunching. The two-ion curve was calculated under the assumption that the ions acted independently.

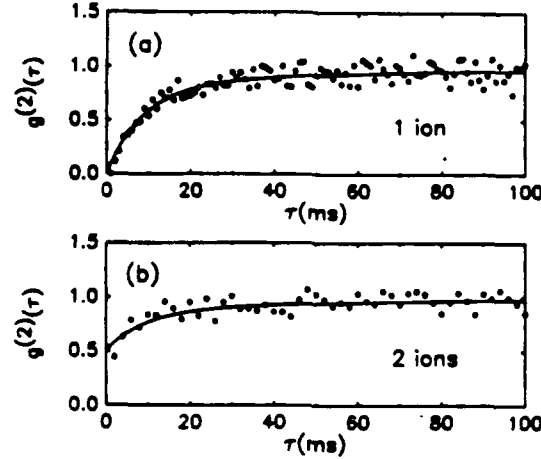


Figure 3: Plots of $g^{(2)}(\tau)$ for (a) one Hg^+ ion and (b) two Hg^+ ions. The dots are experimental data. The solid curves are calculated.

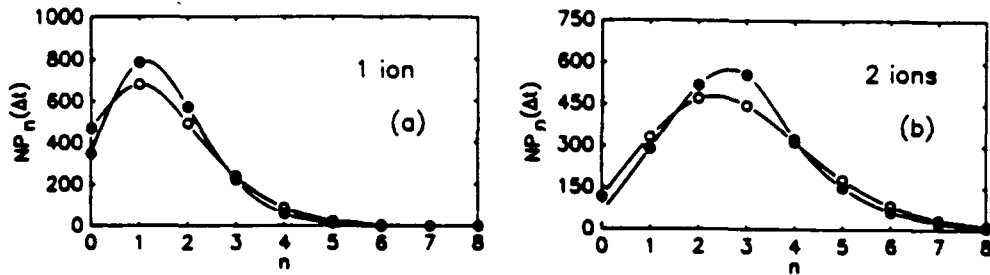


Figure 4. Plots of the distributions of the numbers of $11\ \mu\text{m}$ photons emitted in 200 ms intervals from (a) one Hg^+ ion and (b) two Hg^+ ions. The filled circles are experimental data. The open circles are Poissonian distributions with the same means as the experimental distributions.

Let $P_n(\Delta t)$ be the probability of detecting $n\ 11\ \mu\text{m}$ photons in a time interval Δt . Such a distribution is called sub-Poissonian if its standard deviation σ is less than its mean $\langle n \rangle$. Mandel's Q parameter [$Q = (\sigma^2 - \langle n \rangle) / \langle n \rangle$], a measure of the deviation from Poissonian statistics, was measured to be -0.253 ± 0.025

for one ion and -0.242 ± 0.025 for two ions. The negative values of Q are an indication of sub-Poissonian statistics. Figure 4 shows $NP_n(\Delta t)$ for (a) one ion and (b) two ions, for $\Delta t = 200$ ms and a total measurement time T of 400 s. Here, $N = T/\delta T$ is the total number of time intervals, so that $NP_n(\Delta t)$ is the number of intervals in which n photons were detected. For comparison, Poissonian distributions with the same $\langle n \rangle$ are also shown. This work and related statistical analyses are detailed in refs 1 and 3 of "Papers submitted."

(4.) Lifetimes and Branching Ratios in Hg^+ .

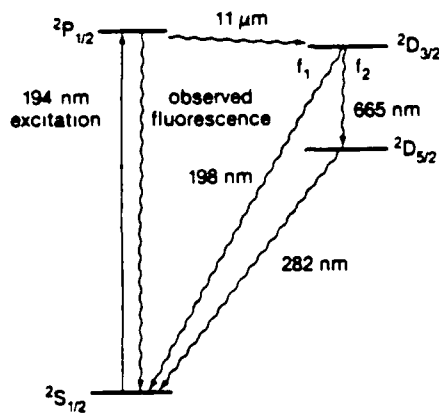


Figure 5. Diagram of the lowest four energy levels of Hg^+ . The decay rates from all three excited levels were determined by observation of only the 194-nm fluorescence.

The radiative decay rates of the $^2D_{3/2}$ level and the $^2D_{5/2}$ level in Hg^+ are reflected in the probability distribution W_{off} of the durations of the "off" quantum jump periods for a single ion irradiated by 194 nm radiation. A calculation based on the rate equations for the probabilities of being in the various levels yields

$$W_{off}(\tau) \propto [f_2 \gamma_2 \exp(-\gamma_2 \tau) + (f_1 \gamma_1 - \gamma_2) \exp(-\gamma_1 \tau)]. \quad (1)$$

Here γ_1 and γ_2 are the total radiative decay rates of the $^2D_{3/2}$ level and the $^2D_{5/2}$ level respectively and the branching ratios f_1 and f_2 are shown in Fig. 5.

The experimental fluorescence-off distribution was least-squares fitted to eq. (1) to obtain values for γ_1 , γ_2 , and f_1 . Figure 6 shows the data and the least-squares fit. The values obtained from the fit are $\gamma_1 = 109 \pm 5 \text{ s}^{-1}$, $\gamma_2 = 11.6 \pm 0.4 \text{ s}^{-1}$, and $f_1 = 0.491 \pm 0.015$. These values are in reasonable agreement with calculations. The value of γ_2 is in good agreement with previous measurements but the value of γ_1 is about a factor of 2 higher than the only previously reported value. No previous measurements of f_1 exist. The measurements are described in more detail in ref. 7 of "Papers Published."

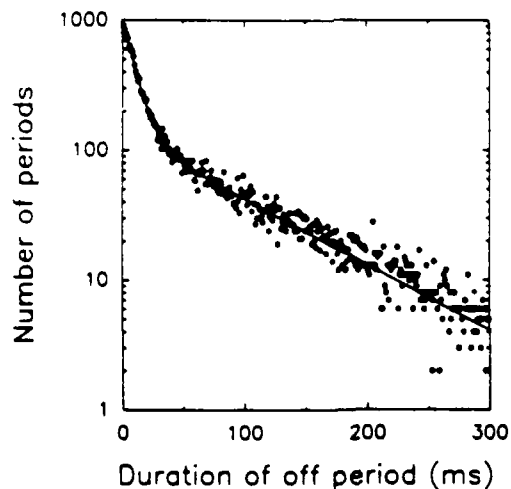


Figure 6. Distribution of fluorescence-off periods for a single ion (dots) and a least-squares fit (curve). These data represent an analysis of approximately 40000 quantum jumps (up or down).

(5.) Laser Cooling in Penning Traps.

Previously, we have experimentally and theoretically investigated laser cooling of single ions in Penning traps. The cooling of many ions is complicated by increased rotation frequencies from space charge, Doppler broadening larger than the natural width, and external torques (due for example to field asymmetries) which can compensate the torque applied by the laser. A new theory has been developed which includes these features. By using numerical techniques, the steady-state temperature of an ion plasma in a Penning trap, cooled by a laser beam perpendicular to the trap axis, is calculated theoretically. The calculated temperature is a function of the intensity, frequency, and position of the laser beam and of the rotation frequency of the plasma. Temperatures of ${}^9\text{Be}^+$ plasmas were measured for a wide range of experimental parameters. The measured and calculated temperatures were in agreement. The lowest and highest temperatures were approximately 40 mK and 2K. The results of these calculations and experiments are discussed in Ref. 4 of "Papers submitted."

(6.) Laser Development. The radiative linewidth of the ${}^2S_{1/2} \rightarrow {}^2D_{5/2}$ transition in Hg^+ (Fig. 5) is approximately 1.8 Hz. This transition, which we anticipate using as the reference for an optical clock, requires a stable narrow linewidth laser at 281 nm. Even though we have made the highest resolution optical measurements on ions so far, we are still limited to about 30 kHz resolution (at 281 nm) because the dye laser drifts in the time required to probe the ion. (The "fast" linewidths of the laser, i.e., neglecting drift, are only about 300 Hz). We can significantly reduce this problem by using an intrinsically more stable laser and by locking the laser to a more stable reference cavity.

As a first step, a Nd:FAP laser has been made to oscillate single frequency at 1.126 μm . This solid state laser (Nd:FAP kindly supplied by R. Byer, Standard Univ.) is intrinsically more stable than a dye laser because dye jet fluctuations are avoided. By doubling this frequency twice to reach 281 nm we anticipate having a much more stable oscillator to probe the quadrupole transition in Hg^+ .

Experience on this type of laser has been gained in similar experiments on Nd:YAG. These experiments are reported in Refs. 5 and 6 of "Papers submitted."

(7.) Coulomb Clusters, Liquid and Solid Plasmas

With the low temperatures achieved by laser cooling, we expect the ions in "clouds" to show spatial correlations including liquid and solid like behavior. This behavior has wide interest in the general physics community because it is seen or expected to be seen in many diverse circumstances including: 2-D configurations of electrons or ions near the surface of liquid helium, charged particles in liquid suspension which act through a shielded Coulomb potential, electrons in solids (Wigner crystallization), astrophysics, in high energy storage rings, and in plasmas.

In addition to this basic physics interest, our work on this subject has grown because the primary systematic frequency shift in spectroscopy on clouds of ion's is still expected to be the second order Doppler frequency shift from the ions residual motion after laser cooling. Therefore a knowledge of the ion's distribution functions is critical for the spectroscopy.

In one experiment, small numbers of $^{198}\text{Hg}^+$ ions were stored in a miniature Paul trap, which has properties equivalent to those of a hyperbolic trap with dimensions $r_0 \approx 466 \mu\text{m}$ and $z_0 \approx 330 \mu\text{m}$. The ions were laser cooled by 1-2 μW of cw 194-nm radiation (bandwidth $< 2 \text{ MHz}$), whose beam waist was varied between 5 and 15 μm at the position of the ions. This radiation was tuned near the $5d^{10}6s \ ^2S_{1/2} \rightarrow 5d^{10}6p \ ^2P_{1/2}$ first resonance line, which has a natural linewidth of 70 MHz. Some of the 194-nm fluorescence from the ions was focused onto the photocathode of a resistive-anode photon-counting imaging tube as shown schematically in Fig. 7. The positions of the photons detected by the imaging tube could be displayed on an oscilloscope in real time or stored by a computer in order to make time exposures.

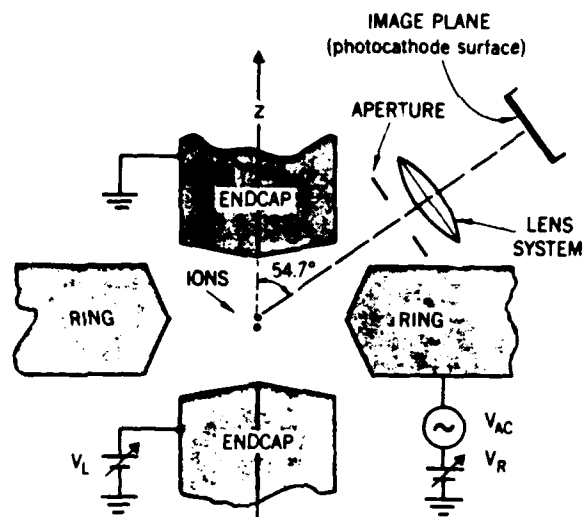


Figure 7. Schematic drawing of the trap electrodes (to scale) and imaging system (not to scale) for Hg^+ cluster experiments. The end-cap to end-cap separation along the z axis is approximately $625 \mu\text{m}$. The overall magnification of the lens system is $180\times$. The laser beam used to illuminate the ions also enters the trap at an angle of 54.7° with respect to the z axis and is perpendicular to the observation axis shown.

In Fig. 8, we show images obtained for a few cases of up to 16 ions. We can characterize the ponderomotive trapping potential by the single ion resonance frequencies ν_z and ν_r for the motion in the axial and radial directions respectively. (In Fig. 8, we assume $\nu_x \approx \nu_y = \nu_r$).

When ν_z/ν_r is made large enough, all of the ions are forced by the strength of the potential in the z direction to lie in the x - y plane as in Figs. 8b and d. When ν_z/ν_r is small enough, the ions lie along the z axis as in Fig. 8c. This allowed us to count the ions. Below the images in Figs. 4, d, e, f, and g we show the configurations obtained theoretically for each value of the number of ions N_i obtained by minimizing the function

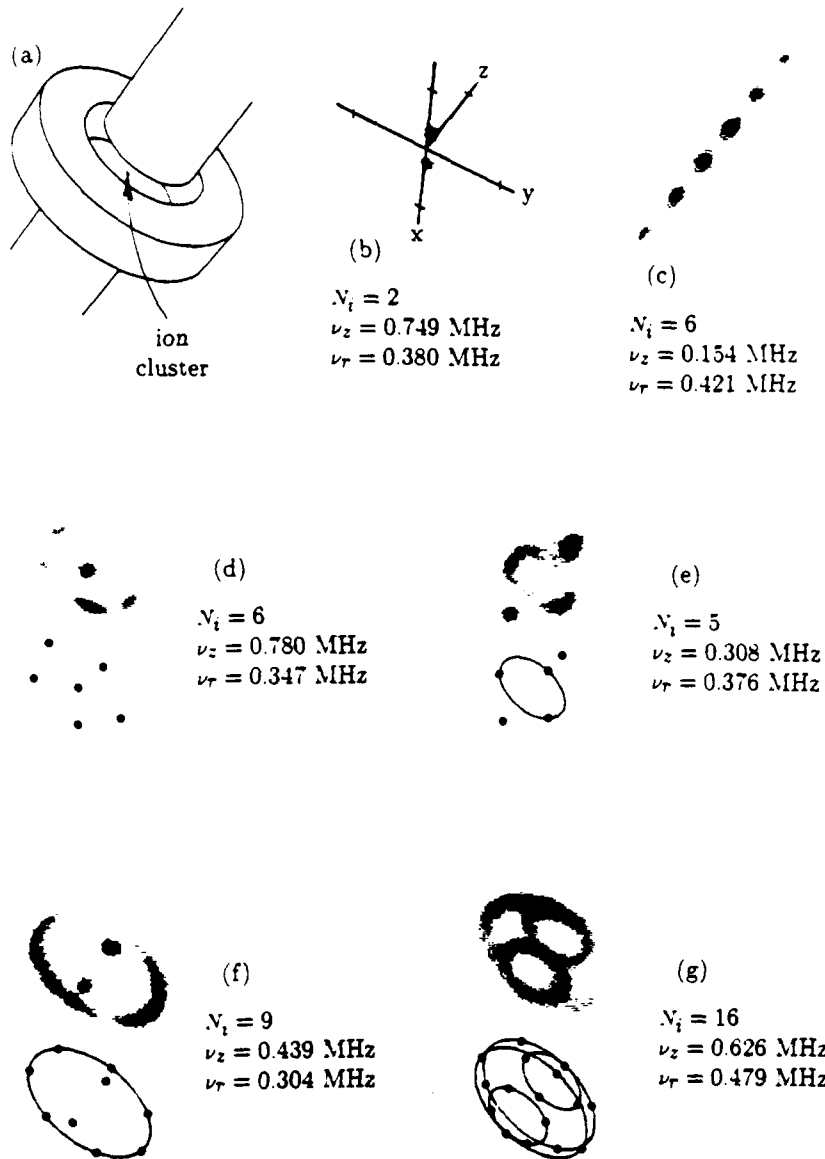


Figure 8. Images of Hg^+ ion "clusters" in a Paul trap. In (a), the trap electrodes are shown schematically in the same orientation as for the remainder of the pictures, but at reduced magnification (inner diameter of the ring electrode is $\approx 0.9 \text{ mm}$). The coordinate system in (b) applies to the rest of the images. In (d), (e), (f), and (g), we display numerical solutions for the expected cluster shapes for the experimental values of ν_z and ν_r . The "rings" are caused by ion circulation about the trap z axis of symmetry. In (d), a non-fluorescing impurity ion occupies a particular ion site in the cluster.

$$E_{\phi} = q \sum_{i=1}^{N_i} (\alpha r_i^2 + \beta z_i^2) + q^2 \sum_{i < j}^{N_i} |\vec{r}_i - \vec{r}_j|^{-1}$$

where α and β are determined from the experimentally measured values of ν_r and ν_z . The circular images or "rings" in Figs. 8e through g are due to ion circulation about the z axis.

In principle, it should be possible to observe ordered structures of much larger numbers of atomic ions in Paul traps. In our trap, this was apparently prevented for $N_i \geq 20$ because the small amount of power available for cooling was unable to overcome the rf heating. In rf heating, the kinetic energy of the ion's driven motion at frequency Ω is coupled into the secular motion characteristic of the pseudopotential well.

Note that in Fig. 8g, we see that 16 ions approximately lie on 3 rings. The larger ring, containing 8 ions, lies in the x-y plane and the smaller rings, each containing 4 ions, lie in planes above and below but parallel to the x-y plane. We may also view these ions as lying on the surface of a spheroidal shell characteristic of the structure for large numbers of ions.

We can also perform optical spectroscopy on these clusters in which case they can be viewed as pseudomolecules. These experiments are unique partially because we can observe each atom in the "molecule" separately. (See Fig. 9)

In Penning-type traps, where ions are confined by static electric and magnetic fields, rf heating does not take place and large collections of ions can be laser-cooled to temperatures less than 10 mK. Several molecular dynamics simulations on collections of a hundred to a few thousand ions confined in storage rings and traps show that when $\Gamma > 1$, the ions are predicted to reside in concentric shells. In "clouds" of laser-cooled $^9\text{Be}^+$ ions confined in a Penning trap we have observed structures containing from 1 shell (≈ 20 ions) to 16 shells ($\approx 15,000$ ions).

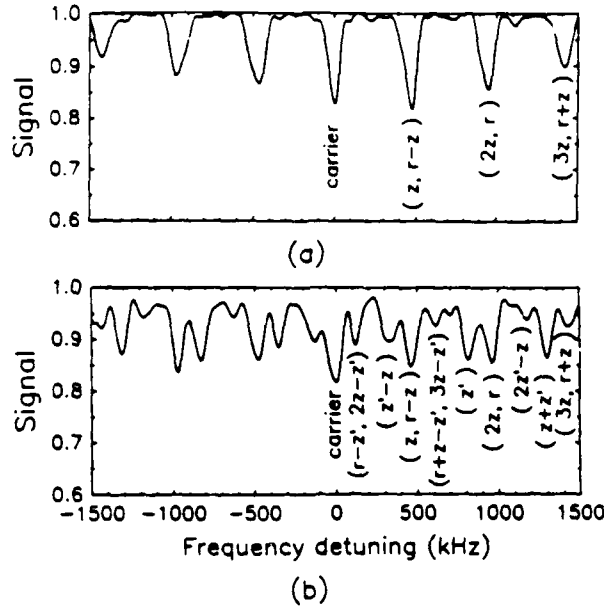


Figure 9. Absorption spectra of the electric quadrupole transition $^2S_{1/2}(m_j=+1/2) \rightarrow ^2D_{3/2}(m_j=1/2)$ for individual Hg^+ ions. (a) The central part of the spectrum for a single trapped ion. Adjacent to the carrier are Doppler-effect-generated sidebands due to the harmonic motion of the ion in the trap. Trapping conditions were adjusted to satisfy $\nu_z \approx 2\nu_z \approx 473$ kHz. The identifications below the lines show the order of the sideband. For example, the identification $(z, r-z)$ denotes absorption at the frequencies $\nu_0 + \nu_z$ and $\nu_0 + \nu_z - \nu_z$, where $\nu_0 \approx 1.07 \times 10^{15}$ Hz is the carrier frequency. (b) The quadrupole spectrum for one of two ions, trapped along the z axis. The trapping conditions were the same as for (a). The new lines are due to the stretch vibration mode of the ions at frequency $\nu_z' \approx \sqrt{3}\nu_z$.

$^9\text{Be}^+$ ions are trapped in the cylindrical Penning trap shown schematically in Fig. 10. A magnetic field $\vec{B} = B\hat{z}$ ($B = 1.92$ T) produced by a superconducting magnet confined the ions in the direction perpendicular to the z axis. A static potential U_0 between the end and central cylinders confined the ions in the z direction to a region near the center of the trap.

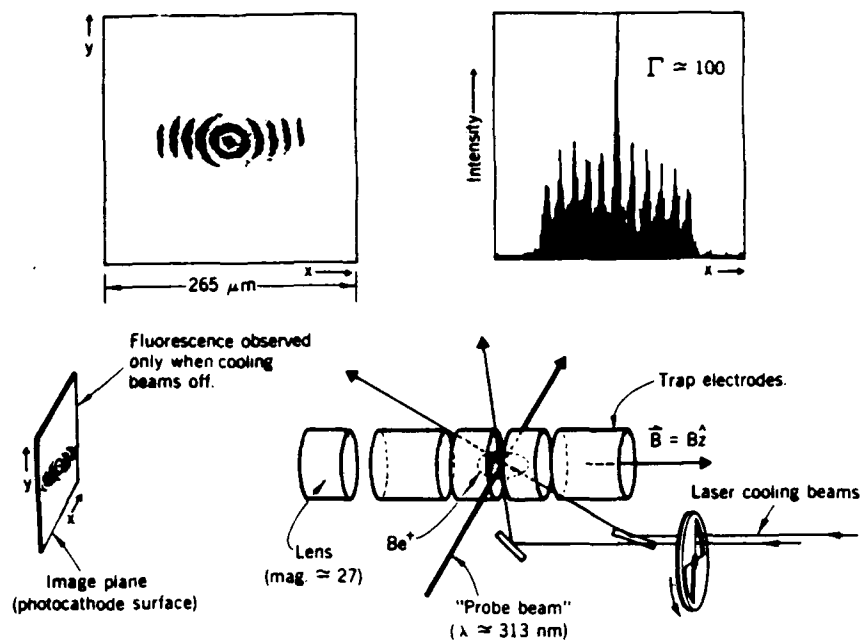


Figure 10. Schematic representation of apparatus for observation of shell structure of strongly coupled ${}^9\text{Be}^+$ ions in a Penning trap. Images are made of the ions which are intersected by the probe beam. In the upper right, an intensity plot vs x for a value of y intersecting the center of the cloud is shown. Shell structure results from the boundary conditions near the edge of the cloud.

The ${}^9\text{Be}^+$ ions were laser cooled by driving the $2s\ {}^2S_{1/2} (m_l = 3/2, m_j = 1/2) \rightarrow 2p\ {}^2P_{3/2} (3/2, 3/2)$ transition slightly below the resonant frequency. The 313 nm cooling radiation ($\approx 30\ \mu\text{W}$) could be directed perpendicular to the magnetic field and/or along a diagonal as indicated in Fig. 10. A second laser (power $\approx 1\ \mu\text{W}$, beam waist $\approx 30\ \mu\text{m}$) was used to spatially map the shell structure of

the cloud. This probe laser was tuned to the same transition as the cooling laser and was directed through the cloud perpendicularly to the magnetic field. With the probe laser on continuously, the cooling laser could be chopped at 1 kHz (50% duty cycle) and the image signal integrated only when the cooling laser was off as shown in Fig. 10. At present, it is difficult to make further quantitative comparisons between our data and the theoretical calculations. For example, there is substantial uncertainty in our measurement of Γ due to uncertainty in the temperature measurement. Also, the spatial resolution is partially limited by optics. Our data do agree qualitatively with the simulations with the exception of the presence, in some cases, of an open cylinder shell structure as opposed to the predicted closed spheroids. These experiments are discussed further in refs. 8 and 9 of "Papers published."

(8) Synchrotron Frequency Divider. A first design for a trap using a metalized ceramic construction which would make an integral trap/vacuum system has been abandoned because of the difficulty of obtaining reliable ceramic seals and the problem of disassembly for modifications. A design using demountable flange is now complete and will be constructed shortly. The superconducting magnet for this experiment has been made operational. The graduate student on this project has recently finished classes and passed his preliminary exams in April, 1988.

(9) Sympathetic cooling. Using Be^+ and Mg^+ ions in the superconducting magnet system, sympathetic cooling has been achieved. Future experiments are detailed in the proposed work section.

OFFICE OF NAVAL RESEARCH

PUBLICATIONS[†] / PATENTS / PRESENTATIONS / HONORS REPORT

1 October 1987 through 30 September 1988

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"COOLED ION FREQUENCY STANDARD"

Principal Investigator

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PAPERS PUBLISHED IN REFEREED JOURNALS

1. "Laser Cooling Limits and Single Ion Spectroscopy," D.J. Wineland, W.M. Itano, J.C. Bergquist and R.G. Hulet, Phys. Rev. A 36, 2220 (1987).
2. "Recoilless Optical Absorption and Doppler Sidebands of a Single Trapped Ion," J.C. Bergquist, W.M. Itano and D.J. Wineland, Phys. Rev. A 36, 428 (1987).
3. "Laser Spectroscopy of Trapped Atomic Ions," W.M. Itano, J.C. Bergquist, and D.J. Wineland, Science 237, 612 (1987).
4. "Quantum Jumps via Spontaneous Raman Scattering," R.G. Hulet and D.J. Wineland, Phys. Rev. A 36, 2758 (1987).
5. "Precise Test of Quantum Jump Theory," R.G. Hulet, D.J. Wineland, J.C. Bergquist, and W.M. Itano, Phys. Rev. A 37, 4544 (1988).
6. "Radiative Decay Rates in Hg^+ from Observations of Quantum Jumps in a Single Ion," W.M. Itano, J.C. Bergquist, R.G. Hulet, D.J. Wineland, Phys. Rev. Lett. 59, 2732 (1987).
7. "Atomic Ion Coulomb Clusters in an Ion Trap," D.J. Wineland, J.C. Bergquist, W.M. Itano, J.J. Bollinger and C.H. Manney, Phys. Rev. Lett. 59, 2935 (1987).
8. "Shell-structure Phase of Magnetically Confined Strongly Coupled Plasmas," S.L. Gilbert, J.J. Bollinger, and D.J. Wineland, Phys. Rev. Lett. 60, 2022 (1988).
9. "Precise Optical Spectroscopy with Ion Traps," W.M. Itano, J.C. Bergquist, R.G. Hulet, and D.J. Wineland, Physica Scripta, T22, 79 (1988).
10. "Cooling in Traps," R. Blatt, G. Lafyatis, W.D. Phillips, S. Stenholm, and D.J. Wineland, Physica Scripta T22, 216 (1988).
11. "Static Properties of a Nonneutral $^9\text{Be}^+$ Ion Plasma," L.R. Brewer, J.D. Prestage, J.J. Bollinger, W.M. Itano, D.J. Larson, and D.J. Wineland, Phys. Rev. A 38, 859 (1988).
12. "Photon Antibunching and Sub-Poissonian Statistics from Quantum Jumps in One and Two Atoms," W.M. Itano, J.C. Bergquist, and D.J. Wineland, Phys. Rev. A, A38, 559 (1988).

PAPERS SUBMITTED TO REFEREED JOURNALS (not yet published)

1. "Perpendicular Laser Cooling of Ion Plasmas in a Penning Trap,"
W.M. Itano, L.R. Brewer, D.J. Larson, and D.J. Wineland,
submitted for publication.
2. "Thermal Shifts of the Spectral Lines in the $^4F_{3/2}$ to $^4I_{11/2}$
Manifold of a Nd:YAG Laser," S. Z. Xing and J.C. Bergquist,
IEEE J. Quant. Electronics, to be published.
3. "Intracavity Doubling in a Single Frequency cw ND:YAG Ring
Laser," S.Z. Xing and J.C. Bergquist, submitted for
publication.
4. "Laser Cooling to the Zero Point Energy of Motion," F.
Diedrich, J.C. Bergquist, W.M. Itano, and D.J. Wineland,
submitted for publication.

BOOKS (and sections thereof) PUBLISHED

1. "The Observation of Quantum Jumps in Hg^+ ," W.M. Itano, J.C. Bergquist, R.G. Hulet, and D.J. Wineland, Laser Spectroscopy VIII, W. Persson and S. Svanberg eds., (Springer Verlag, Berlin, Heidelberg, 1987) p. 117.
2. "Ion Traps for Large Storage Capacity," D.J. Wineland, Proc. of the Cooling, Condensation, and Storage of Hydrogen Cluster Ions Workshop, SRI International, Menlo Park, CA Jan., 8, 9, 1987, editor J.T. Bahns, p. 181.

BOOKS (and sections thereof) SUBMITTED.

1. "Ion Trapping Techniques: Laser Cooling and Sympathetic Cooling," J.J. Bollinger, L.R. Brewer, J.C. Bergquist, W.M. Itano, D.J. Larson, S.L. Gilbert, and D.J. Wineland, Proc. 1987 Workshop on Intense Positron Beams, submitted for publication.
2. "Liquid and Solid Ion Plasmas," D.J. Wineland, W.M. Itano, J.C. Bergquist, S.L. Gilbert, J.J. Bollinger, and F. Ascarunz, Proc. ONR Nonneutral Plasma Physics Symposium, Washington, D.C., submitted for publication.
3. "Quantum Optics Experiments with a Single Ion," R.G. Hulet, J.C. Bergquist, W.M. Itano, and D.J. Wineland, Proc. Int. Laser Science Meeting, Baltimore, Oct. 1987.
4. "Frequency Standards in the Optical Spectrum," D.J. Wineland, J.C. Bergquist, W.M. Itano, F. Diedrich and C.S. Weimer, in The Hydrogen Atom, Ed. by F. Bassani, T.W. Hänsch and M. Inguscio, (Springer Verlag, Heidelberg, 1988) to be published.
5. "The Digitized Atom and Optical Pumping," D.J. Wineland, W.M. Itano, J.C. Bergquist and R.G. Hulet, in Atomic Physics 11, ed.

by S. Haroche, J.C. Gay, G. Grynberg, (World Scientific Press, Singapore, 1988) to be published.

6. "Liquid and Solid Phases of Laser Cooled Ions," S.L. Gilbert, J.C. Bergquist, J.J. Bollinger, W.M. Itano, and D.J. Wineland, in Atomic Physics 11, (World Scientific Press, Singapore, 1988) to be published.
7. " Hg^+ Single Ion Spectroscopy," J.C. Bergquist, F. Diedrich, W.M. Itano, and D.J. Wineland, Proc. 4th Symp. on Frequency Standards and Metrology, Ancona, Italy (Springer Verlag, 1988) to be published.
8. "High Accuracy Spectroscopy of Stored Ions," D.J. Wineland, W.M. Itano, J.S. Bergquist, J.J. Bollinger, F. Diedrich and S.L. Gilbert, Proc. 4th Symp. on Frequency Standards and Metrology, Ancona, Italy (Springer Verlag, 1988) to be published.
9. "Frequency Standards Utilizing Penning Traps," J.J. Bollinger, S.L. Gilbert, W.M. Itano, and D.J. Wineland, Proc. 4th Symp. on Frequency Standards and Metrology, Ancona, Italy (Springer Verlag, 1988) to be published.
10. "Quantative Study of Laser Cooling in a Penning Trap," W.M. Itano, L.R. Brewer, D.J. Larson, J.J. Bollinger, and S.L. Gilbert, and D.J. Wineland, Proc. 4th Symp. on Frequency Standards and Metrology, Ancona, Italy (Springer Verlag, 1988) to be published.
11. "Laser Cooling," D.J. Wineland, McGraw - Hill Encyclopedia of Science and Technology, to be published.
12. "Atomic Clocks," W.M. Itano, McGraw - Hill Encyclopedia of Science and Technology, to be published.

PATENTS FILED

None

PATENTS GRANTED

None

INVITED PRESENTATIONS AT TOPICAL OR SCIENTIFIC/TECHNICAL SOCIETY CONFERENCES
(June 1987 to September 1988)

1. "Liquid and Solid Plasmas," Gordon Conf. on Atomic Physics, July '87; J.J. Bollinger.
2. "Precision Spectroscopy Using Trapped Ions," Symposium on the Physics of Stored and Trapped Particles, Sweden, June '87; W.M. Itano.
3. "Cooling Limits in Traps," Symposium on the Physics of Stored and Trapped Particles, Sweden, June '87; D.J. Wineland.
4. "The Observation of Quantum Jumps in Hg^+ ," 8th Conf. on Laser Spectroscopy, Sweden, June '87; W.M. Itano.
5. "Strongly Coupled Nonneutral Ion Plasmas," '87 Conf. on Atomic Processes in Plasmas, Santa Fe, NM, September '87; J.J. Bollinger.
6. "Sympathetic Cooling of Positrons by Atomic Ions," Intense Positrons Beam Workshop, Idaho Falls, June '87; J.J. Bollinger.
7. "Spectroscopy of a Single Trapped Ion," Rocky Mountain Section of Opt. Soc. Am., September '87; J.C. Bergquist.
8. "Quantum Optics Experiments with a Single Ion," International Laser Science Meeting, Baltimore, October '87; R.G. Hulet.
9. "Ordered Structures of Cold, Stored Ions," Div. Atomic, Molecular, and Optical Physics, APS, Baltimore, April '88, J.J. Bollinger.
10. "Liquid and Solid Behavior of Strongly Coupled Plasmas," Workshop on New Directions in Plasma Engineering, Berkeley, June '88; J.J. Bollinger.
11. "Single Ion Optical Frequency Standard," Conf. on Precision Electromagnetic Measurements, Japan, June '88; J.C. Bergquist.
12. "Liquid and Solid Phases of Laser Cooled Ions," Int. Conf. on Atomic Physics, Paris, July '88; S.L. Gilbert.
13. "Frequency Standards in the Optical Spectrum," Symposium on The Hydrogen Atom, Pisa, July '88; D.J. Wineland.
14. "The Digital Atom and Optical Pumping," Symposium in honor of Jean Brossel, Paris, July '88; D.J. Wineland.
15. "Laser Cooling (and Freezing) of Stored Ions," Ann. Meeting of AAPT, Cornell, June '88; J.C. Bergquist.

16. "Liquid and Solid Ion Plasmas," ONR Nonneutral Plasma Physics Symposium, Nat. Acad. of Sciences, March '88; D.J. Wineland.
17. "Experiments with Laser-Cooled, Trapped Ions," Adriatico Research Conf. on Frontier Sources for Frontier Spectroscopy, Pisa, August '88; J.J. Bollinger.
18. "Penning Trap Frequency Metrology," Fourth Symposium on Frequency Standards and Metrology, Ancona, Italy, September '88; J.J. Bollinger.
19. " Hg^+ Single Ion Spectroscopy," Fourth Symposium on Frequency Standards and Metrology, Ancona, Italy, September '88; J.C. Bergquist.
20. "Generation of Ultra-Stable cw Optical Sources," Adriatico Research Conf. on Frontier Sources for Frontier Spectroscopy," Pisa, Italy, August '88; J.C. Bergquist.
21. "Ion Frequency Standards," Fourth Symposium on Frequency Standards and Metrology, Ancona, Italy, September '88; D.J. Wineland.

OTHER INVITED TALKS (Colloquia, etc.)

1. "Laser Cooling and Spectroscopy of Stored Atomic Ions," Colorado School of Mines, September '87; S.L. Gilbert.
2. "Single Atom Spectroscopy," Univ. of Missouri, Rolla, September '87; D.J. Wineland.
3. "Spectroscopy of a Single Trapped Ion," Argonne National Lab., October '87; J.C. Bergquist.
4. "Spectroscopy of a Single Trapped Ion," Notre Dame Univ., October '87; J.C. Bergquist.
5. "High Resolution Spectroscopy of a Single, Laser-Cooled Ion," NBS, Gaithersburg, October '87; J.C. Bergquist.
6. "Quantum Jumps, Antibunching, and Spectroscopy of a Single Ion," Stanford Univ., December '87; J.C. Bergquist.
7. "Characteristics of Laser-Cooled Ion Plasmas Confined in a Penning Trap," Univ. of California, San Diego, December '87; S.L. Gilbert.
8. "Strongly Coupled Stored Ions; Coulomb Clusters, Liquid and Solid Plasmas," Los Alamos National Lab., March '88; D.J. Wineland.
9. "Shell Structure in Strongly Coupled Ion Clouds. A Smectic Phase?" NBS, Gaithersburg, April '88; S.L. Gilbert.
10. "Laser Cooling (and Freezing) of Stored Ions," Univ. of Calif., Santa Cruz, April '88; J.C. Bergquist.
"Ion Crystals, Large and Small," MIT, Cambridge, MA, April '88; D.J. Wineland.
"Spectroscopy of Single Ions," Univ. Nevada, Las Vegas, May '88; D.J. Wineland.
13. " Strongly Coupled Stored Ions; Coulomb Clusters Liquid and Solid Plasmas," Univ. of Washington, May '88; D.J. Wineland.
14. "Applications of Laser Cooled Ions," Bell Labs, Murray Hill, NJ; May '88, J.J. Bollinger.
15. "Spectroscopy of Laser Cooled Ions," Institute Laue-Langevin, Grenoble France, July '88, W.M. Itano.
16. "Spectroscopy of Laser Cooled Ions," Univ. of Texas, Austin, September '88, D.J. Wineland.

HONORS/AWARDS/PRIZES

1. Election to Fellow, National Bureau of Standards: D.J. Wineland

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